Spin transport in graphene-based van der Waals heterostructures

Ingla Aynés, Josep

Chapter 10

Conclusions and outlook

Abstract

This chapter presents the conclusions of this thesis and compares the results described here with those from other groups. A short outlook is provided about open questions and future potential perspectives related with each topic addressed in this thesis.

10.1 High-quality graphene for long distance spin transport

The first results on hBN-encapsulated graphene devices in 2014 showed spin relaxation lengths up to 12 µm with 2 ns spin lifetimes at room temperature [1], more than a factor of two longer than the previous achieved values for suspended graphene [2] and graphene on hBN [3]. The use of a double gated geometry, which allowed for independent control of the carrier density and electric field, allowed to show that the spin lifetime can be tuned with the electric field [1]. Motivated by these findings and the results on bilayer graphene on SiO_2 obtained in [4, 5], we studied spin transport in hBN-encapsulated bilayer graphene that lead to the observation of spin relaxation lengths up to 24 µm with 3 ns lifetimes at 5 K and 13 µm with 1.2 ns at room temperature, as shown in Chapter 5. These promising results represent a lower bound to the spin relaxation length in the studied devices. As our calculations show, the spin lifetimes are limited by the non-encapsulated regions where the ferromagnetic contacts are placed. Hence, to achieve longer spin relaxation lengths and determine the intrinsic limits of such devices one has to fabricate samples with encapsulated regions long enough so that the encapsulated regions dominate the spin relaxation.

Another approach to achieve high quality graphene devices for spintronics is the use of few layer hBN as a tunnel barrier for efficient spin injection and a protection barrier to protect the graphene from the fabrication induced residues. The first results were achieved in monolayer hBN [6, 7], however, this approach has proven very successful when using bilayer and trilayer hBN barriers, showing large, bias dependent contact polarizations and spin lifetimes in the ns range[8–12].

In parallel with the encapsulated graphene project developed in our lab, the group from Aachen developed a technique to transfer hBN/graphene heterostructures on ferromagnetic cobalt contacts with MgO barriers [13] leading to comparable results as in [1]. Further improvements and the use of large and thick enough hBN protective layers have lead to the observation of spin lifetimes up to 12 ns in monolayer graphene using this approach [14], currently the longest achieved spin lifetime in a graphene spin valve.

10.2 Spin guiding using drift currents

Typical spin transport experiments are carried out using spin diffusion. This process is slow and, hence, is not the most optimal way to perform long distance spin transport. To increase the distance over which one can transport spin information, we studied the effect of drift on spin transport in high quality bilayer graphene. Our results, shown in Chapter 6, show that the spin relaxation length increases up to 90 μm and most of the spins (88% in our device) can be guided with the drift.

The drift control of the spin propagation offers new possibilities for different device applications. A recently introduced device geometry for spin currents is the gate controlled spin current demultiplexer [15]. In this geometry, the spin current in a graphene bifurcation is guided to the desired output by tuning the spin resistance ($R_{sq}\lambda/W$) of the different arms. The spins diffuse in the arm which has the lower spin resistance that can be changed by electrostatic gating. However, the efficiency of such guiding is quite limited by the fact that the spin relaxation length decreases as the channel resistance increases, limiting the contrast between the ′on′ and ′off′ arms. To increase the guiding efficiency we have developed a new way to perform such operations using spin drift in the different arms. Our results from Chapter 7 show that, by changing the carriers from electrons to holes in the different arms it is possible to guide the spin currents with efficiencies more than an order of magnitude higher than in the diffusive experiment with moderate drift current densities of 20 μA/μm.

Another advantage of the spin drift approach is that, since drift occurs much faster than diffusion, it can be used for the fast device operations required to realize practical applications. The major drawback of the spin drift approach is the power consumption that arises from the drift current application. This consumption needs to be reduced further to achieve useful operations. We propose the use of 2D semiconductors such as black phosphorous, where the spin lifetimes are in the range of nanoseconds [16]. Its semiconducting nature allows for the carrier densities to be reduced down to orders of magnitude lower than in monolayer graphene at room temperature. Because the drift velocity is inversely proportional to the carrier density, the drift currents required to achieve efficient operation can be greatly reduced [17].

Another approach to realize spin-based logic using carrier drift is the use of graphene pn junctions. The output in this case is the spin accumulation instead of the spin current like in the demultiplexer. In this case, because the drift velocity is opposite for electrons and holes, the spin accumulation at the pn junction gets amplified when the drift velocities at the left and right sides point towards the junction [18]. This is because diffusion away from the junction is greatly reduced by the effect of drift. This geometry also allows for logic operations since the spin accumulation at the pn junction can be efficiently tuned by the drift.

10.3 Proximity induced spin orbit coupling in TMD/graphene heterostructures

The ability to fabricate heterostructures with different layered materials and the 2D nature of graphene allow the control of the electronic properties of graphene via proximity to different materials [19]. The control of spin-orbit coupling is crucial to tune the spin relaxation [20] and realize logic operations using spin currents. With this purpose, we studied spin transport in graphene in close proximity with a monolayer of transition metal dichalcogenide posessing a large spin-orbit coupling. By studying spin precession around an in-plane magnetic field we determined that the in-plane spin lifetime is of 3.5 ps and the out-of-plane one of 40 ps, as shown in Chapter 8 [21], in agreement with the theoretical predictions from [22] and experimental results obtained using a tilted B field technique [23]. These results are a signature of the proximity effect imprinted on the graphene layer.

A promising route towards the achievement of new spintronic applications using graphene/TMD heterostructures is to induce proximity on double-gated bilayer graphene. When a perpendicularly applied electric field induces a bandgap, the bands become layer polarized and this allows for the tunning of the spin-orbit coupling in double gated bilayer graphene [24, 25].

However, there is another effect which is relevant for the study of graphene/TMD heterostructures, which is the spin absorption. Since the in-plane spin lifetimes in the TMD layer are shorter than in graphene, activation of the TMD conductivity leads to a flow of spins that relax in the TMD layer. This provides a way to control the spin signal across the TMD/graphene heterostructure with a gate voltage[26, 27].

10.4 Anisotropic spin transport in bilayer graphene

The biggest puzzle in graphene spintronics is the source of spin relaxation [28]. A useful tool to determine its origin is to study the spin lifetime anisotropy. Anisotropy has been studied using high magnetic fields (above 1 T) that pull the contact magnetizations out of the plane and allow to compare between the nonlocal signals generated by in-plane and out-of-plane spins [29]. However, this technique only allows for accurate determination of the anisotropy at high carrier densities where the effect of magnetoresistance is negligible [1]. Recently, a technique applying magnetic fields at different angles with respect to the injected spins and extracting the non-precessing spin component has been developed [30]. This procedure does not require high magnetic fields and it is suitable to determine the spin lifetime anisotropy at the charge neutrality point. In monolayer graphene spin relaxation is mostly isotropic [30] and the anisotropy can be reduced with perpendicularly applied electric fields that induce in-plane Rashba fields [1]. However, in bilayer graphene, it has been reported theoretically that the intrinsic spin-orbit fields induce a large spin lifetime anisotropy near the charge neutrality point [31, 32].

In Chapter 9 we have studied the spin lifetime anisotropy in bilayer graphene using tilted magnetic fields and obtained an anisotropy of 8 at the charge neutrality point with out-of plane spin lifetimes up to 9 ns and in-plane lifetimes up to 1.8 ns. These results are the first demonstration of spin lifetime anisotropy in bilayer graphene, are consistent with the theoretical predictions, and represent the first experimental result that indicates that intrinsic spin-orbit fields dominate spin relaxation in a graphene-like system. Moreover, our results indicate that resonant scattering by magnetic impurities does not limit the spin lifetime in our device since that would lead to isotropic spin relaxation [33]. This anisotropy has also been shown recently to change with the perpendicular electric field [34].

References

[1] M. Guimarães, P. Zomer, J. Ingla-Aynés, J. Brant, N. Tombros, and B. J. van Wees, "Controlling spin relaxation in hexagonal BN-encapsulated graphene with a transverse electric field," *Physical Review Letters* **113**(8), 086602, (2014).

[2] M. H. Guimarães, A. Veligura, P. Zomer, T. Maassen, I. Vera-Marun, N. Tombros, and B. J. van Wees, "Spin transport in high-quality suspended graphene devices," *Nano Letters* **12**(7), 3512, (2012).

[3] P. Zomer, M. Guimaraes, N. Tombros, and B. J. van Wees, "Long-distance spin transport in high-mobility graphene on hexagonal boron nitride," *Physical Review B* **86**(16), 161416, (2012).

[4] W. Han and R. K. Kawakami, "Spin relaxation in single-layer and bilayer graphene," *Physical Review Letters* **107**(4), 047207, (2011).

[5] T.-Y. Yang, J. Balakrishnan, F. Volmer, A. Avsar, M. Jaiswal, J. Samm, S. Ali, A. Pachoud, M. Zeng, M. Popinciuc, *et al.*, "Observation of long spin-relaxation times in bilayer graphene at room temperature," *Physical Review Letters* **107**(4), 047206, (2011).

[6] T. Yamaguchi, Y. Inoue, S. Masubuchi, S. Morikawa, M. Onuki, K. Watanabe, T. Taniguchi, R. Moriya, and T. Machida, "Electrical spin injection into graphene through monolayer hexagonal boron nitride," *Applied Physics Express* **6**(7), 073001, (2013).

[7] M. V. Kamalakar, A. Dankert, J. Bergsten, T. Ive, and S. P. Dash, "Enhanced tunnel spin injection into graphene using chemical vapor deposited hexagonal boron nitride," *Scientific Reports* **4**, 6146, (2014).

[8] M. V. Kamalakar, A. Dankert, P. J. Kelly, and S. P. Dash, "Inversion of spin signal and spin filtering in ferromagnet— hexagonal boron nitride-graphene van der waals heterostructures," *Scientific Reports* **6**, 21168, (2016).

[9] M. Gurram, S. Omar, S. Zihlmann, P. Makk, C. Schönenberger, and B. J. van Wees, "Spin transport in fully hexagonal boron nitride encapsulated graphene," *Physical Review B* **93**(11), 115441, (2016).

[10] S. Singh, J. Katoch, J. Xu, C. Tan, T. Zhu, W. Amamou, J. Hone, and R. Kawakami, "Nanosecond spin relaxation times in single layer graphene spin valves with hexagonal boron nitride tunnel barriers," *Applied Physics Letters* **109**(12), 122411, (2016).

[11] M. Gurram, S. Omar, and B. J. van Wees, "Bias induced up to 100% spin-injection and detection polarizations in ferromagnet/bilayer-hBN/graphene/hBN heterostructures," *Nature Communications* **8**(1), 248, (2017).

[12] M. Gurram, S. Omar, S. Zihlmann, P. Makk, Q. Li, Y. Zhang, C. Schönenberger, and B. J. van Wees, "Spin transport in two-layer-cvd-hBN/graphene/hBN heterostructures," *Physical Review B* **97**(4), 045411, (2018).

[13] M. Drögeler, F. Volmer, M. Wolter, B. Terrés, K. Watanabe, T. Taniguchi, G. Güntherodt, C. Stampfer, and B. Beschoten, "Nanosecond spin lifetimes in single-and few-layer graphene–hBN heterostructures at room temperature," *Nano Letters* **14**(11), 6050, (2014).

[14] M. Drögeler, C. Franzen, F. Volmer, T. Pohlmann, L. Banszerus, M. Wolter, K. Watanabe, T. Taniguchi, C. Stampfer, and B. Beschoten, "Spin lifetimes exceeding 12 ns in graphene nonlocal spin valve devices," *Nano Letters* **16**(6), 3533, (2016).

[15] X. Lin, L. Su, Z. Si, Y. Zhang, A. Bournel, Y. Zhang, J.-O. Klein, A. Fert, and W. Zhao, "Gate-driven pure spin current in graphene," *Physical Review Applied* **8**(3), 034006, (2017).

[16] A. Avsar, J. Y. Tan, M. Kurpas, M. Gmitra, K. Watanabe, T. Taniguchi, J. Fabian, and B. Özyilmaz, "Gate-tunable black phosphorus spin valve with nanosecond spin lifetimes," *Nature Physics* **13**(9), 4141, (2017).

[17] J. Kikkawa and D. Awschalom, "Lateral drag of spin coherence in gallium arsenide," *Nature* **397**(6715), 139, (1999).

[18] I. J. vera-Marun, V. Ranjan, and B. J. van Wees, "Nonlinear interaction," *Physical Review B* **84**(24), 241408, (2011).

[19] A. K. Geim and I. V. Grigorieva, "Van der Waals heterostructures," *Nature* **499**(7459), 419, (2013).

[20] S. Datta and B. Das, "Electronic analog of the electro-optic modulator," *Applied Physics Letters* **56**(7), 665, (1990).

[21] T. S. Ghiasi, J. Ingla-Aynés, A. A. Kaverzin, and B. J. van Wees, "Large proximity-induced spin lifetime anisotropy in transition-metal dichalcogenide/graphene heterostructures," *Nano Letters* **17**(12), 7528, (2017).

[22] A. W. Cummings, J. H. Garcia, J. Fabian, and S. Roche, "Giant spin lifetime anisotropy in graphene induced by proximity effects," *Physical Review Letters* **119**(20), 206601, (2017).

[23] L. A. Benítez, J. F. Sierra, W. S. Torres, A. Arrighi, F. Bonell, M. V. Costache, and S. O. Valenzuela, "Strongly anisotropic spin relaxation in graphene–transition metal dichalcogenide heterostructures at room temperature," *Nature Physics* **14**(3), 303, (2018).

[24] M. Gmitra and J. Fabian, "Proximity effects in bilayer graphene on monolayer WSe_2: Field-effect spin valley locking, spin-orbit valve, and spin transistor," *Physical Review Letters* **119**(14), 146401, (2017).

[25] J. Y. Khoo, A. F. Morpurgo, and L. Levitov, "On-demand spin–orbit interaction from which-layer tunability in bilayer graphene," *Nano Letters* **17**(11), 7003, (2017).

[26] W. Yan, O. Txoperena, R. Llopis, H. Dery, L. E. Hueso, and F. Casanova, "A two-dimensional spin field-effect switch," *Nature Communications* **7**, 13372, (2016).

[27] A. Dankert and S. P. Dash, "Electrical gate control of spin current in van der Waals heterostructures at room temperature," *Nature Communications* **8**, 16093, (2017).

[28] W. Han, R. K. Kawakami, M. Gmitra, and J. Fabian, "Graphene spintronics," *Nature Nanotechnology* **9**(10), 794, (2014).

[29] N. Tombros, S. Tanabe, A. Veligura, C. Jozsa, M. Popinciuc, H. Jonkman, and B. Van Wees, "Anisotropic spin relaxation in graphene," *Physical review letters* **101**(4), 046601, (2008).

[30] B. Raes, J. E. Scheerder, M. V. Costache, F. Bonell, J. F. Sierra, J. Cuppens, J. Van de Vondel, and S. O. Valenzuela, "Determination of the spin-lifetime anisotropy in graphene using oblique spin precession," *Nature Communications* **7**, 11444, (2016).

[31] S. Konschuh, M. Gmitra, D. Kochan, and J. Fabian, "Theory of spin-orbit coupling in bilayer graphene," *Physical Review B* **85**(11), 115423, (2012).

[32] L. Wang and M. Wu, "Electron spin relaxation in bilayer graphene," *Physical Review B* **87**(20), 205416, (2013).

[33] D. Kochan, S. Irmer, M. Gmitra, and J. Fabian, "Resonant scattering by magnetic impurities as a model for spin relaxation in bilayer graphene," *Physical Review Letters* **115**(19), 196601, (2015).

[34] J. Xu, T. Zhu, Y. K. Luo, Y.-M. Lu, and R. K. Kawakami, "Strong and tunable spin-lifetime anisotropy in dual-gated bilayer graphene," *Physical Review Letters* **121**(12), 127703, (2018).